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Fredenslund, Anders Michael; Scheutz, Charlotte

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TOTAL METHANE LOSS FROM BIOGAS PLANTS, DETERMINED BY TRACER DISPERSION MEASUREMENTS

A.M. FREDENSLUND* AND C. SCHEUTZ*

** Department of Environmental Engineering, Technical University of Denmark, Bygningstorvet, Building 115, 2800 Kongens Lyngby, Denmark*

SUMMARY: methane losses from biogas plants are problematic, since methane emitted into the atmosphere contributes to global warming, and any losses may thus reduce the environmental benefits of biogas production. A tracer gas dispersion method was used to measure total methane emissions from seven biogas plants, and more measurement campaigns are planned. Emissions varied between 1.3 and 13.4 (kg CH_4 h^{-1}), and losses expressed in percentages of production varied between 0.3 and 6.4%. The tracer gas dispersion method was found to be useful in quantifying total methane emissions from biogas plants, whereas the detection and quantification of individual leaks at the plants require other methods.

1. INTRODUCTION

The anaerobic digestion of organic waste as a waste management option can provide several environmental advantages compared to other treatment options, such as the opportunity to recycle nutrients (such as nitrogen and phosphorous) and generate a fuel (biogas) that can be utilised for a large number of energy purposes. Another positive environmental impact is the possibility of significant greenhouse gas (GHG) reductions (Møller et al., 2009; Styles et al., 2016; Wulf et al., 2006). GHG accounting procedures for biogas plants often include the substitution of fossil fuels, plant energy use, the substitution of chemical fertilisers, transportation of biomass/digestate and GHG emissions from biogas engines and/or biogas upgrade units. Fugitive emissions from biogas plants are often omitted, and sometimes studies mention that there is a lack of information on the magnitude of these emissions.

Due to the high global warming potential of methane, fugitive emissions are potentially important. Explaining that “the fugitive loss of methane is difficult to establish by measurements and probably highly variable from facility to facility”, Møller et al. (2009) calculated GHGs emission from treating organic waste as being between 0 and 48 kg CO_2-eq. $tonne^{-1}$ wet waste (ww) received at the facility, assuming a methane loss of between 0 and 3%. When taking into account various biogas utilisation options and variations for a number of key parameters, Møller et al. (2009) found that the global warming impact for biogas production from organic waste varied between -375 kg (a net saving) and 111 kg CO_2-eq. $tonne^{-1}$ ww (a net load). This study shows that a methane loss of 3% is significant compared to the overall performance of biogas production from organic waste with regards to GHG mitigation.

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In the case study of a biogas plant planning to receive organic industrial waste, manure and seaweed from a beach cleaning operation, Solrød Biogas was expected to accept 155,000 tonnes of biomass (ww) per year and produce 5.43 million m^3 CH_4 annually (Fredenslund et al., 2011). In this study, the global warming impact of biogas production from the feedstock mix was found to be between -259 kg and -163 kg CO_2-eq. $tonne^{-1}$ ww, depending on the utilisation option (biogas upgrade, process energy at a nearby factory or combined heat and power). Considering the amount of treated waste, the net GHG reduction from the facility was calculated as being between 25,280 tons CO_2-eq. yr^{-1} and 40,140 tons CO_2 eq. yr^{-1}. These figures were reached by considering fossil fuel substitution, biogas plant energy use, the substitution of chemical fertiliser, transportation of biomass/digestate and avoided GHG emissions through a change in manure management, though methane loss was not taken into account. The magnitude of methane lost from the site, which would result in a net GHG reduction of 0, was equivalent of 23% to 37% of total biogas production. Methane losses on this scale are probably very rare, but the numbers indicate, similarly to Møller et al. (2009), that methane losses from biogas production can significantly affect the overall performance of biogas production with regards to GHG mitigation.

To maximise both GHG emission reduction and biogas plant energy production, it is relevant to ensure that methane losses are maintained as low as possible. The detection of leaks, their quantification and practical measures to mitigate them are important tools in meeting this objective. However, on-site measuring methods used to quantify leaks may not provide an accurate assessment in this regard, since quantification requires that leaks are detectable and measurable, and more diffuse emissions may not be taken into account. Thus, the risk of underestimating total fugitive methane emissions from biogas plants (and similar locations) is a weakness of on-site methods that measure leaks individually. Alternatively, remote sensing techniques allow total fugitive emissions to be measured, if these methods are able to monitor the entire gaseous plume emitted from a plant. These methods may be highly useful for regulatory purposes (i.e. monitoring if a plant's emissions are above or below a given threshold) or for other purposes where the total emission is the most relevant figure, such as providing data for environmental assessments.

This paper summarises the preliminary results of several ongoing projects in which a tracer gas dispersion method was applied to quantify total methane emissions from biogas plants.

2. MATERIALS AND METHODS

2.1 Tracer gas dispersion measurements

The tracer gas dispersion methodology described in this paper was originally developed at DTU Environment to measure total methane emissions from landfills (Mønster et al., 2015, 2014). The method is now used to measure total methane emissions from landfills in Denmark in relation to the biocover initiative to minimise such discharges. The method has also been used to study fugitive emissions from other facilities, such as waste water treatment plants (Yoshida et al., 2014) and composting facilities (Andersen et al., 2010). Other research groups have utilised similar methods to quantify fugitive emissions, albeit with some variations in instrumentation and choice of tracer gas (Börjesson et al., 2007; Czepiel et al., 2003; Foster-Wittig et al., 2015).

The tracer gas dispersion method combines the controlled release of a tracer gas from the site from which the emission is to be determined with concentration measurements downwind, using a mobile high-resolution analytical instrument. The basic principle behind

the method is the assumption that a tracer gas released from an emission source will disperse into the atmosphere in the same way as methane emitted from the source. Assuming that the wind direction is defined, conditions in the air above the landfill are sufficiently combined for the methane and tracer gas to be mixed fully, and when the tracer gas release and methane emission are constant, the methane emission rate (E_{gas}) can be calculated as a function of the ratio of the integrated cross-plume concentration of methane emitted into the integrated cross-plume concentration of the tracer gas, according to Eq. 1 (Mønster et al., 2015):

$$E_{gas} = Q_{tracer} \cdot \frac{\int_{Plume\ end\ 1}^{Plume\ end\ 2} C_{gas} dx}{\int_{Plume\ end\ 1}^{Plume\ end\ 2} C_{tracer} dx} \cdot \frac{MW_{gas}}{MW_{tracer}} \quad (1)$$

where Q_{tracer} is the release rate of the tracer gas (kg h^{-1}), C_{gas} and C_{tracer} are concentrations of methane and the tracer (ppbv) above background level, MW_{gas} and MW_{tracer} are the molecular weights of methane and the tracer gas and x is the distance across the plume.

In this study, inorder to place tracer gas in the most accurate location to simulate methane emissions, an initial methane concentration screening was performed at the site, which provided a qualitative assessment of the emission pattern. From this screening, one or more locations were chosen. An adequate simulation of the emission pattern of the site would result in a close match between the methane and tracer gas plumes measured downwind from the site. If an improper source simulation was observed, the placement of the tracer gas release was changed prior to emission quantification. The method is described in more detail in Mønster et al. (2014), which also includes an assessment of the method's measurement accuracy.

2.2 Instrumentation

Concentrations of methane were measured using a cavity ring down spectrometer (G2203, Picarro Inc., USA) installed in a car. Atmospheric gas was pumped into the analyser from an intake placed on the roof of the car and approximately 2 m above ground level. The measurement frequency of the instrument was approximately 2 Hz, and the precision levels of the methane and acetylene measurements were 0.48 ppb and 0.40 ppb, respectively, which enabled the detection of small variations in atmospheric concentrations.

A GNSS system was used to log the position of the measurements (R330 GNSS receiver and A43 antenna, Hemisphere, Canada), whilst a weather station mounted on the vehicle was used to log the temperature and atmospheric pressure (All-In-One weather sensor, model 102780, Climatronics, USA).

The gas tracer used for the measurements in this study was acetylene (C_2H_2), and 21L gas bottles containing 3.2 kg acetylene each were used for continuous tracer release. The release flows from the bottles were controlled using calibrated flow meters and regulators (Sho-Rate, Brooks Instruments, Holland).

2.3 Surveyed biogas plants and measurement campaigns

Total methane emission measurements from seven biogas plants are included in this paper. The biogas plants varied in terms of size, substrates used, biogas utilisation (combined heat and power (CHP) and/or upgrade and gas grid injection) and more. All facilities used continuously stirred reactors (CSTRs). At each biogas plant, methane concentration screening was performed as described in section 2.1. For each measurement

taken to quantify methane emissions while releasing the acetylene tracer gas, 20 cross-plume readings were taken at minimum, and the methane emission was calculated as an average value of the performed measurements. In some cases, individual plume transects were disregarded either due to interference from a nearby methane source, such as a farm, or due to a poor signal-to-noise ratio for the measured concentrations. Details of the measurements and characteristics of the biogas plants are listed in Table 1.

Table 1. Details of the biogas plants and measurement campaigns

Biogas plant	Main substrate(s)	Gas utilisation	Date of measurement	Number of tracer release locations	Number of transects used in calculations
A	Manure, organic waste	CHP	Feb. 18, 2016	2	15
B	Manure, organic waste	CHP	Oct. 26, 2015 and Nov 5. 2015	1	58
C	Manure, energy crops	Biogas upgrade	Nov. 4, 2015	1	28
D	Manure, organic waste	Biogas upgrade	Nov. 12, 2015	1	49
E	Manure, grass silage	CHP	Apr. 4, 2016	2	16
F	Waste water sludge	CHP + biogas upgrade	Apr. 29, 2016	2	18
G	Maize silage	CHP + biogas upgrade	Oct. 24 to Oct. 28, 2016	1 to 3	214

For each measurement, daily biogas production was provided by the biogas plant operator, in order to help determine methane loss relative to production. Biogas plant G is located in Germany, while the remaining biogas plants are located in Denmark.

3. RESULTS AND DISCUSSION

Figure 1 shows two examples of methane and tracer concentrations measured while traversing the downwind plumes. The top figure shows measurements at biogas plant D at two distances, marked as “Measurement A” and “Measurement B” in the figure. As was the case for some of the other plants, a livestock farm was located nearby, which was also a source of methane discharge. In this case, it was possible to distinguish clearly between methane plumes from the biogas plant and farm. The bottom figure shows an example from a measurement at biogas plant G, which illustrates a close correlation between measured methane concentrations above background level and concentrations of the gaseous tracer. This correlation indicates that the tracer gas satisfactorily simulated methane released from the facility, which was the case for all seven biogas plants, probably due to these units being more “focused” sources of emission, where methane is released from a relatively small area compared to other sources such as landfills.

In general, peak concentrations of methane and tracer in the measured plumes were between approximately 4-5 and 200 parts per billion (ppb) above background level. The measurement distances varied between a few hundred metres to more than one kilometre. The measurement distances depended on the availability of drivable roads downwind. This

methodology requires precise instrumentation with regards to determining gas concentrations. The instrument used in this study helped quantify methane emissions from all biogas plants, even though they were relatively low at some facilities.

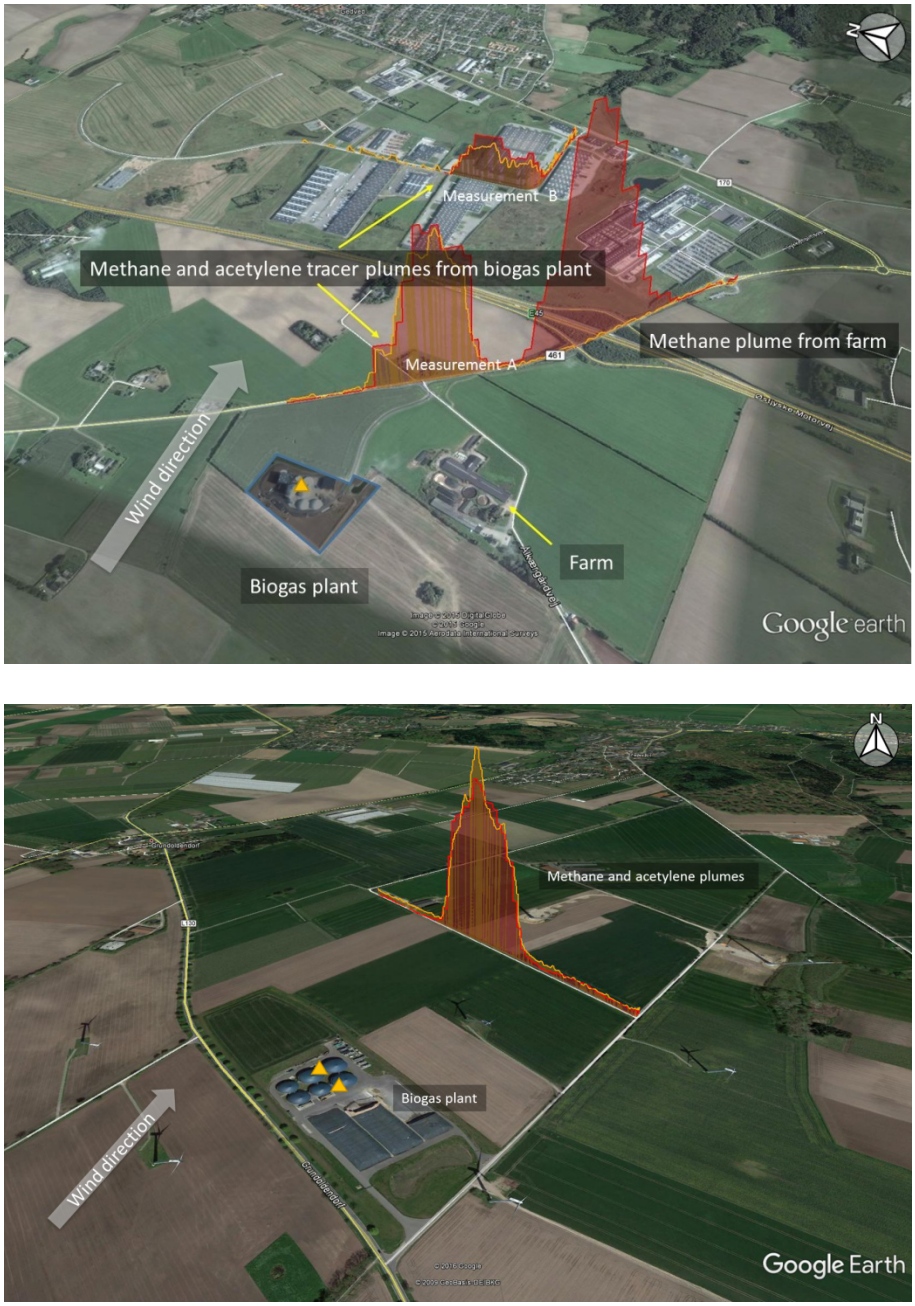

Figure 1. Examples of tracer dispersion measurement at a biogas plants D (top) and G (bottom). The height of the red curve is proportional to the measured concentration of methane above background level, whereas the yellow curve is proportional to the concentration of acetylene tracer gas. The yellow triangles show where the acetylene tracer was released at the facilities.

Table 2. Average biogas production and methane emissions from seven biogas plants

Biogas plant	Biogas production (kg CH_4 h^{-1})	Measured total methane emission (kg CH_4 h^{-1})	Methane loss (%)
A	416	1.3	0.3
B	404	6.4	1.6
C	406	13.4	3.3
D	525	12.6	2.4
E	64	4.1	6.4
F	99	5.0	5.0
G	400	2.1	0.5
All plants (sum)	2314	45.0	1.9

Measured methane emissions varied between 1.3 and 13.4 (kg CH_4 h^{-1}) (see Table 2). At biogas plant D, we were informed that the biogas upgrade system was overloaded and was thus expected to emit more methane than under normal operating conditions; therefore, we plan to repeat our measurements at that facility in due course. Methane loss (emission/production) varied between 0.3 and 6.4%. The lowest emission was measured at a relatively large and recently constructed biogas plant with off-site gas utilisation (CHP), and it was therefore expected that methane loss would be low. The numbers suggest that it is possible to operate biogas plants with methane losses below one per cent, albeit significantly higher emissions have been recorded at some facilities. Summing the gas production and measured methane emissions from the seven biogas plants, we found that total emissions equated to 1.9% of total production. More measurement campaigns are planned to expand the dataset and to evaluate variations in emissions by repeating measurements at the same locations.

4. CONCLUSIONS

A tracer gas dispersion method was used to measure total methane emissions from seven biogas plants, and more measurement campaigns are planned. Methane loss from the plants varied between 0.3 and 6.4% of production. The tracer gas dispersion method was found to be useful in quantifying total methane emissions from these sources; however, the detection and quantification of individual leaks require the use of other measurement methods.

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